Detection of Spiral photons in Quantum Optics

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We show that a new type of photon detector, sensitive to the gradients of electromagnetic fields, should be a useful tool to characterize the quantum properties of spatially-dependent optical fields. As a simple detector of such a kind, we propose using magnetic dipole or electric quadrupole transitions in atoms or molecules and apply it to the detection of spiral photons in Laguerre-Gauss (LG) beams. We show that LG beams are not true hollow beams, due to the presence of magnetic fields and gradients of electric fields on beam axis. This approach paves the way to an analysis at the quantum level of the spatial structure and angular momentum properties of singular light beams.

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Presently Glauber's theory is widely used to describe the quantum properties of optical fields and the detection of photons. In [1], he justifies that in optics, one can restrict in most cases to a detector only sensitive to the electric field amplitude. The electric dipole (E1) detector he considers, is assumed to be of negligible size and extra wide frequency band (electric dipole approximation). Such an assumption will be shown to be restrictive when the spatial structure of optical fields becomes very complicated. As an example of such complicated fields one should mention complicated optical fields near nanostructures. Another example of optical fields with complex space structure is the Laguerre-Gauss beams with phase singularity and zero electric field on axis.They are also called spiral beams, and have attracted a lot of interest owing to their orbital angular momentum[2,3].

The full quantum description of optical fields in such cases is a very complicated problem, and the development of efficient sensors and photon detectors in such fields is a very actual task. To characterize such quantum fields we suggest to use instead of usual E1 detectors, detectors which are sensitive to gradients of electric fields, or to magnetic fields.

The utility of gradient detectors is already proved in hydroacoustic [4], where combined receivers, i.e. devices consisting of scalar sound pressure sensors and several velocity receivers (with mutually perpendicular axes) are widely used to increase sonar antennas efficiency.

Here, we consider as a click of the detector a specfic atomic excitation (or the observation of a deexcitation process, such as a fluorescence on a strong line collected over all space). The specific nature of the detector is that the atomic exciation is reached through a magnetic dipole or electric quadrupole transitions

The excitation probability for the most general detector can be found from usual Fermi's golden rule. In the general case the excitation probability can be expressed through series of gradients of Green function of exciting quantum field [5]. In the case of coherent narrow band optical fields, one gets a simpler quasiclassical expression:

$$\begin{aligned} R_{i\to f} &= \frac{\left|T^{if}\right|^2}{\hbar^2\sqrt{\delta\omega^2+\Gamma^2/4}} \\ T^{if} &= \mathbf{d}^{if}\mathbf{E}\left(\mathbf{r},\omega_0\right)+\mathbf{m}^{if}\mathbf{B}\left(\mathbf{r},\omega_0\right)+Q_{ij}\nabla_i E_j\left(\mathbf{r},\omega_0\right)+... \end{aligned} \tag{1}$$

where $\mathbf{d}^{if},\mathbf{m}^{if},Q_{ij}$ are matrix elements of electric dipole ($E1$), magnetic dipole($M1$) and electric quadrupole ($E2$)transitions between ground and excited states respectively and where $\delta\omega$ and Γ stand for characteristic excitation detuning and transition linewidth respectively. We also assume that the orientation of the detector (molecule) is fixed in space and no additional averaging is needed. Although there can be many Zeeman sublevels in the detector's excited state, only some of them are of interest for detecting complex optical fields. For example there can be 3 magnetic dipole (m^M, $M=-1,0,1$) and 5 quadrupole transitions (Q^M, $M=-2,-1,0,1,2$) the matrix elements of which can be parameterized in the following form within Cartesian co-ordinates (x,y,z) where quantization axis z is chosen along the axis of the light beam:

$$\mathbf{m}^{\pm1}=m^{(1)}\left(\pm1,i,0\right),\mathbf{m}^{0}=m^{(0)}\left(0,0,\sqrt{2}\right)$$

$$\mathbf{Q}^{(0)}=Q^{(0)}\sqrt{\tfrac{2}{3}}\begin{Vmatrix} -1 & 0 & 0\\ 0 & -1 & 0\\ 0 & 0 & 2\end{Vmatrix}$$

$$\mathbf{Q}^{(\pm1)}=Q^{(1)}\begin{Vmatrix} 0 & 0 & \mp1\\ 0 & 0 & -i\\ \mp1 & -i & 0\end{Vmatrix}$$

$$\mathbf{Q}^{(\pm2)}=Q^{(2)}\begin{Vmatrix} 1 & \pm i & 0\\ \pm i & -1 & 0\\ 0 & 0 & 0\end{Vmatrix} \tag{2}$$

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Usually the main contribution to detector interaction with light is due to $(E1)$ transitions (justifying Glauber's ideal photon detector [1]). However such transitions give no contribution to excitation of molecules in regions where there is no electric field! $(M1)$ and $(E2)$ transitions give the predominant contribution to excitation rate in this situation. Let us consider this important case in more details for spiral LG beams.

In the case of LG beams,the electric field can be represented by the following formulae [2,3]

$$\mathbf{E}^m(\mathbf{r},\omega) = E_0 \frac{w_0}{k}\left\{k\alpha U, k\beta U, i\left(\alpha\frac{\partial U}{\partial x}+\beta\frac{\partial U}{\partial y}\right)\right\} e^{ikz} \tag{3}$$

with

$$U = \frac{C_p^{|m|}}{w(z)}\left[\frac{\sqrt{2}r}{w(z)}\right]^{|m|}\exp\left(-\frac{r^2}{w^2(z)}\right)L_p^{|m|}\left(\frac{2r^2}{w^2(z)}\right)\times \exp\left(\frac{ikr^2 z}{2\left(z^2+z_R^2\right)} - im\varphi - i\left(2p+|m|+1\right)\arctan\left(z/z_R\right)\right) \tag{4}$$

where (r,φ,z) are cylidrical coordinates, E_0 is the amplitude of electric field,$C_p^{|m|} = \sqrt{2p!/\pi\left(p+|m|\right)!}$ is the normalization constant, $w(z) = w_0\sqrt{1+z^2/z_R^2}$ is the beam radius at z, $w_0$is the Gaussian beam waist, $L_p^{|m|}(x)$ is the generalized Laguerre polynomial, and $z_R = kw_0^2/2$ is the Rayleigh range of the beam. $p+1$ gives the number of nodes of the field in the radial direction.

The most important properties of Laguerre-Gauss beams is that they can carry both spin and orbital angular momentum, and the total momentum per photon is given by the formula

$$j_z = \hbar\left(m+\sigma\right), \text{with} \quad \sigma = -i\left(\alpha\beta^* - \beta\alpha^*\right) \tag{5}$$

where $m\hbar$ is the orbital angular momentum carried by the beam along its propagation direction [2,3].

The properties of this orbital angular momentum carried by a photon have been mostly addressed at the macroscopic level, the only experiment performed to date [6] at the quantum level having attracted much attention due to the opening of new sets of variable in entanglement. Davila Romero et al. [7] have recently presented the quantized version of this field. Our contribution here is to add to this quantum theory the proposal of detectors for the quanta created in the LG basis.

Below for distinctness we will consider LG beams with $\alpha = 1/\sqrt{2}, \beta = i/\sqrt{2}$ case. It corresponds to LG beam with spin equal to -1 (circular polarization). So the total angular momentum of our LG beam is described by $j_z = \hbar\left(m-1\right)$.

Another key feature of nontrivial $(|m|>1)$ LG beams is the zero of electric energy density of (4) on beam axis. Due to this fact LG beams are often referred to as hollow beams or "doughnut beams" because the electric field vanishes on the axis.

However we find that magnetic energy density and gradients of electric field are nonzero at the axis [5]. For

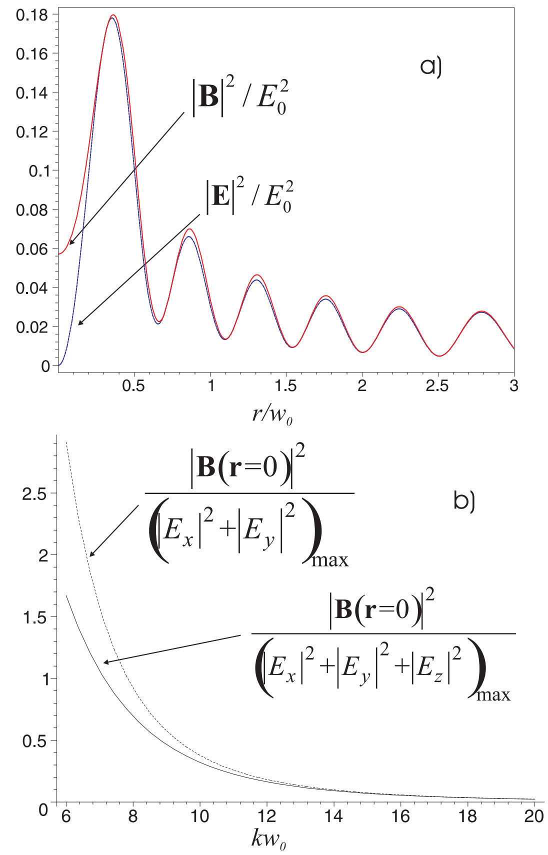

FIG. 1: (a) Radial dependence of the electric and magnetic energy density of LG beam in the waist plane $(kw_0 = 10, p = 6, m = 2)$(b) Ratio of the magnetic energy density at the center of the LG beam to the electric energy density at its maximum as function of beam waist $kw_0(p = 6, m = 2)$, as calculated in the waist plane $z = 0$. For a strong focusing, the elementary $(kw_0)^{-4}$ dependence (dotted line) no longer holds because of a nonnegligible longitudinal component E_z

example, for LG beams with $m = 2$, the magnetic energy density, I_M, on the axis is:

$$I_M = \frac{c\,|B|^2}{8\pi} = \frac{cE_0^2}{8\pi}\frac{32\left(p+1\right)\left(p+2\right)}{\pi\left(kw_0\right)^4} \tag{6}$$

Figure 1 shows that, for strongly focused beams, the magnetic energy on axis becomes comparable to the electric energy density at its maximum.

The nonzero value of magnetic energy at the beam axis is not an artefact of the paraxial nature of LG beam [5].It appears for any general nonparaxial form of monochromatic beam with near cylindrical symmetry [5]

From a formal point of view (Faraday's law of electromagnetic induction), non zero radial magnetic fields on beam axis are due to the presence of longitudinal electric fields in the beam. These longitudinal electric fields are more intense for more focussed beams and for more zeroes in the radial direction. Deeper insight in this matter shows that nonzero on-axis magnetic (or electric) fields

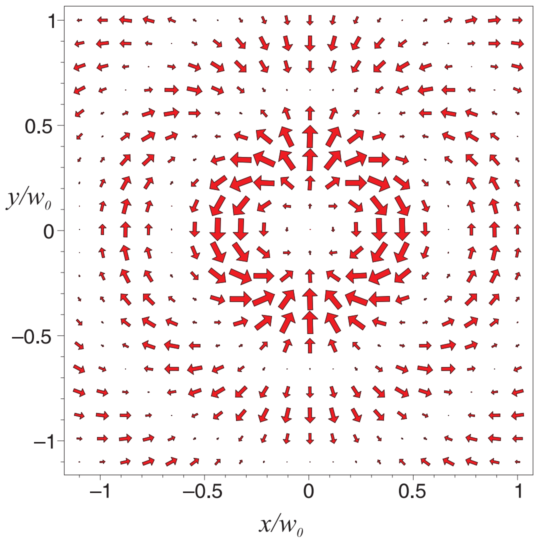

FIG. 2: Space distribution in the waist plane of the real part of the electric field in a Laguerre-Gauss beam($kw_0 = 6, p = 6, m = 2$). The distribution rotates at the optical frequency

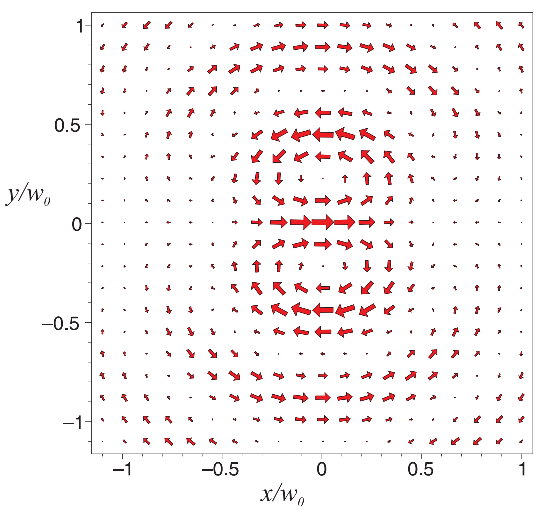

FIG. 3: Space distribution in the waist plane of the real part of the magnetic field in a Laguerre-Gauss beam($kw_0 = 6, p = 6, m = 2$). The distribution rotates at the optical frequency

are related with difficulties in defining a unique phase for the vector fields.

The phase-space structure of Laguerre-Gauss beam is very complicated in comparison with usual circular polarized light. From Figure 2,3 showing the distributions of electric and magnetic field at the waist plane the complicated magnetic structure of LG beams is evident.

The most interesting feature is that both magnetic field (Figure 3) and gradients of electric fields are nonzero at axis so that E1 photon detector cannot work here. Also even in the case of a linear polarization of electric fields, magnetic fields will rotate in space with optical frequency.

Obviously very interesting effects can occur with this electromagnetic energy lying in the region that makes the beam hollow. To test these effects we suggest using new type of detectors described above. As an example we have calculated the quadrupole transition amplitudes $\left(T_Q^{mM} = Q_{ij}^M \nabla_i E_j^m \left(\mathbf{r} = 0\right), m, M = -2, -1, 0, 1, 2\right)$ of a molecule placed on the axis of LG beam :

$$T_Q^{-1,-2} = \frac{4i\sqrt{2p+2}}{\sqrt{\pi}w_0} E_0 Q^{(2)} \tag{7a}$$

$$T_Q^{0,-1} = \frac{2\left(8p+4-(kw_0)^2\right)}{kw_0^2\sqrt{\pi}} E_0 Q^{(1)} \tag{7b}$$

$$T_Q^{1,0} = i\frac{4\sqrt{p+1}}{\sqrt{3\pi}} \frac{\left(8p+8-3\,(kw_0)^2\right)}{k^2 w_0^3} E_0 Q^{(0)} \tag{7c}$$

$$T_Q^{2,1} = \frac{8\sqrt{(p+1)(p+2)}}{\sqrt{\pi}kw_0^2} E_0 Q^{(1)} \tag{7d}$$

Other elements of $T_Q^{m,M}$vanish.

Eqs. (7a-7d) show that the excitation amplitudes are not symmetric under (m,M)->(-m,-M) transformation. This is due to interplay between orbital and spin momenta. As a result, by analyzing (7) one can concludes that such a beam has an angular momentum $j_z = \hbar\,(m-1)$ and $\sigma = -1$,which is in agreement with the independent calculations of angular momentum of the beam (Eq.5) and conservation of angular momentum. An analogous situation takes place for magnetic($M1$) part of transition amplitudes [5].

The most striking feature is the interaction of $m = 2$ LG beam with $M = 1$ transitions in molecules. On axis there is no interaction with E1 transition, while there is an efficient interaction with $E2$ and $M1$ transitions. The on-axis excitation rate for such transitions can be written in the form:

$$R = E_0^2 \frac{64\,(p+1)\,(p+2)}{\hbar^2\sqrt{\delta\omega^2 + \Gamma^2/4\pi}\,(kw_0)^4} \left| m^{(1)} + kQ^{(1)} \right|^2 \tag{8}$$

where $m^{(1)}$ and $Q^{(1)}$ are scalar amplitudes of magnetic dipole and electric qudrupole transitions (see (2)).The spatial distribution of this excitation rates, shown in fig. 4 , is also of great interest.

One sees that this excitation rate has a well pronounced maximum at the beam axis. This property should allow exciting selectively molecules located near the axis, with a sub-wavelength resolution reminiscent of confocal microscopy. Also, this indicates that a photon can be transfered on-axis, in spite of the common idea that a LG beam is a hollow beam. It is very important that a $2\hbar$ exchange of angular momentum is possible in a single atom-photon transition in contrast to a situation for an $E1$ transition [8]

For such experiments, an issue is certainly the low oscillator strength of E2 or M1 transitions, usually considered as nearly forbidden transitions. However, our

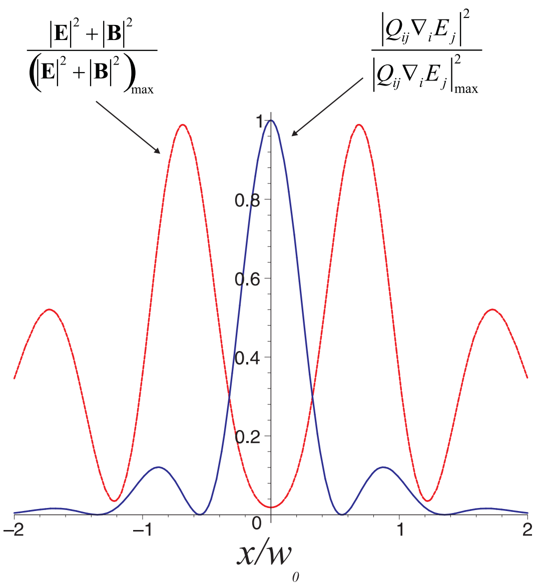

FIG. 4: Normalized radial distribution of excitation rates of an $E2(M=1)$ transitions of an atom placed in the waist plane of LG beams with *p=1,m=2,* $kw_0 = 10$.Dashed line shows normalized distribution of full energy density. An identical distribution is found for an $M1$ transition .

scheme clearly offers the possibility of detecting processes for which a Glauber-type detector is blind. Note that the feasibility of detecting an E2 transition with an evanescent wave (i.e. another type of e.m. field with a complicated structure [9]) was recently established [10]. In the case of LG beams, a strong focusing, and a large value of p, enable the specific contribution associated to $m = 2$ (see eq. 7d) to be comparable to the common plane-wave contribution (eq. 7b) originating in the longitudinal field gradient. The lower sensitivity of $E2$ or $M1$ transition, relatively to $E1$ transition used in ideal dipole detector, is due to the small size of electronic orbit, relatively to optical wavelength (one has typically $m^{(1)} \sim kQ^{(2)} \sim kea^2$, where a- characteristic size of molecules used in detector, while for E1, $d^{(1)} \sim ea$). Hence, stronger non-E1 mechanisms are to be expected with long molecules (e.g. twisted or bio-molecules). A fascinating possibility apparently offered with LG beams is the selective manipulation of chiral molecules (the optical activity is usually associated to a coupled $E1 - M1$ transition) deposited somewhere close to the hollow region. Although a negative experimental result was obtained in [11], our investigation of the local properties of the e.m. field suggests that it is a too large spatial averaging that has made the effect unobservable.

In conclusion, in this letter, we suggest to use new type of detectors, which are sensitive to gradients of electric fields and to magnetic fields, in order to detect photons with a complicated space structure. We have applied our idea to LG beams for which we have shown that spiral beams should not be considered as hollow, because of nonzero magnetic fields on the axis of the beam for orbital momentum number $m = 2$. Our direct calculation of excitation rates of such transitions confirm that Laguerre-Gauss beams bear angular momentum, that can be transferred in an elementary exchange with a quantum system, hence relaxing the usual selection rules. Despite we consider coherent state of exciting fields our conclusions and proposals are valid for any quantum state of exciting field.

From a quantum optics point of view, the investigation of the nature of spiral photons generated with LG beams has remained until now extremely limited [6]. Indeed, all experimental investigations involving LG beams and their specific angular momentum (see [2] and also [6,12,13]) have been integrated on at least a micron-size volume, instead of using a negligible size detector. If particle physics considerations had shown that electromagnetic fields can bear a large angular momentum [14], the coherent production of large number of identical spiral photons in the optical domain is a only recent achievement, susceptible to open new frontiers in quantum optics (e.g. quantum limits to spatial correlation,...). Our suggestion of using a detector that is not E1-type, but whose size remains intrinsically microscopic, should help to elucidate experimentally the quantum properties of these photons carried by a LG or a singular beam, whose specificity appears enhanced under a strong focusing according to our semiclassical deriveation. This regime of sharply focused propagating beams also opens a natural connection with the domain of nano-optics, where it is known that the relative strength of $E2$ transition is enhanced [15]. More generally, with the development of nanotechnologies, it becomes conceivable to produce suitable non-E1 detectors, such as an artificial nanoparticle of special shape (nanoantennas) designed to be sensitive to gradients of electric fields. In return, these detectors should benefit to the very contemporary characterization of more complicated nanooptical fields worth being tested.

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